



Ultra-high resolution mass separator—Application to detection of nuclear weapons tests

K. Peräjärvi^{a,*}, T. Eronen^b, V.-V. Elomaa^b, J. Hakala^b, A. Jokinen^b, H. Kettunen^b, V.S. Kolhinen^b, M. Laitinen^b, I.D. Moore^b, H. Penttilä^b, J. Rissanen^b, A. Saastamoinen^b, H. Toivonen^a, J. Turunen^a, J. Äystö^b

^a STUK-Radiation and Nuclear Safety Authority, P.O. Box 14, FI-00881 Helsinki, Finland

^b Department of Physics, University of Jyväskylä, P.O. Box 35 (YFL), FI-40014 University of Jyväskylä, Finland

ARTICLE INFO

Article history:

Received 4 September 2009

Received in revised form

8 October 2009

Accepted 3 December 2009

Keywords:

Comprehensive nuclear-test-ban treaty

Noble gas samplers and spectrometers

Pure ^{133m}Xe sample

Penning trap

ABSTRACT

A Penning trap-based purification process having a resolution of about 1 ppm is reported. In this context, we present for the first time a production method for the most complicated and crucially important nuclear weapons test signature, ^{133m}Xe . These pure xenon samples are required by the Comprehensive Nuclear-Test-Ban Treaty Organization to standardize and calibrate the worldwide network of xenon detectors.

© 2009 Elsevier Ltd. All rights reserved.

1. Introduction

Radioactive noble gases produced in nuclear weapons tests offer a sensitive probe for the verification of the compliance with the Comprehensive Nuclear-Test-Ban Treaty (Bowyer et al., 2002). Even in the case of underground detonation it is probable that noble gases are released into the atmosphere (New Scientist, 2007; Ringbom et al., 2009). In practice, the detection effort is focused on ^{131m}Xe ($T_{1/2}=11.84$ d), ^{133m}Xe ($T_{1/2}=2.19$ d), ^{133}Xe ($T_{1/2}=5.243$ d) and ^{135}Xe ($T_{1/2}=9.14$ h) (Wernsperger and Schlosser, 2004). Their relative amounts in a sample, in particular the ratio of ^{133m}Xe to ^{133}Xe , are key indicators (Reeder and Bowyer, 1998) to reveal the origin of the sample, i.e., to attribute the source to a nuclear test or to releases from nuclear reactors or isotope production plants. The above mentioned xenon isotopes and isomers are ideal for the verification of the Treaty compliance because of their large production cross sections in fission and long enough half-lives (Bowyer et al., 2002). Below we present a technique for producing pure samples of these isotopes and isomers using the most challenging case, ^{133m}Xe , as an example.

In mass, the isomeric and the ground state of ^{133}Xe differ only by about 2 ppm. Such a small mass difference coupled with the identical chemical behavior make the production of pure ^{133m}Xe samples very challenging. None of the presently available

techniques allow the straightforward production of high-purity (> 90%) samples of ^{133m}Xe . For example, the beta decay of ^{133}I is not a useful method of producing highly enriched ^{133m}Xe samples since it mainly (97.1%) feeds the ground state of ^{133}Xe . Neutron-induced fission coupled to rapid chemical manipulation on the other hand is governed by the direct xenon production cross sections. For example, in case of the neutron-induced fission of ^{235}U the initial purity ($^{133m}\text{Xe}/[^{133m}\text{Xe}+^{133}\text{Xe}]$) is around 73% yet ^{135}Xe is simultaneously produced with a yield about 19 × higher in comparison to ^{133m}Xe (Bowyer et al., 2002). Due to its shorter half-life, ^{135}Xe can be removed from the sample via radioactive decay. Extended decay time, however, also significantly worsens the $^{133m}\text{Xe}/[^{133m}\text{Xe}+^{133}\text{Xe}]$ ratio.

Laser-based separation techniques do not work well either because the first excited atomic state of xenon is at too high energy to be reached with existing lasers and DC- or RF-discharge sources used to populate metastable states from which lasers can be utilized are rather inefficient. Storage rings do not possess high enough mass resolving power to separate the isomeric and ground states of ^{133}Xe . A Penning trap is an existing device which has the highest potential for reaching the required resolving power for purification (Brown and Gabrielse, 1986). Using well established trapping techniques in a gas-filled Penning trap (Savard et al., 1991), a mass resolution of 20 ppm is routinely reached. A more accurate cleaning method has been recently demonstrated in the Accelerator Laboratory of the University of Jyväskylä, Finland, reaching 4 ppm for ^{50}Mn and ^{54}Co (Eronen et al., 2008a). For ^{133m}Xe preparation, the resolution had to be

* Corresponding author. Tel.: +358 9 759 88 705; fax: +358 9 759 88 433.
E-mail address: kari.perajarvi@stuk.fi (K. Peräjärvi).

improved by about a factor of two, without sacrificing the throughput.

2. Experimental techniques and results

The desired ^{133m}Xe nuclides were synthesized using proton-induced fission reactions in a natural uranium target (thickness 15 mg/cm²). The proton beam energy was 25 MeV and intensity 5 μA . The synthesized nuclides recoiling out from the uranium target were thermalized in flowing helium gas which subsequently transported them out from the target chamber. A fraction of the recoils exit the target chamber as ions. These ions were separated from the neutral buffer gas and accelerated to 30 keV of kinetic energy by using electric fields and differential pumping. After acceleration, the ion beam was mass separated using a dipole magnet. In the present work the magnet was tuned to transmit only species with a mass-over-charge ratio of 133 prior to further manipulation. For more information about the used Ion Guide Isotope Separator On-Line technique, see Åystö (2001).

The mass-separated ion beam was transferred to a buffer-gas filled radiofrequency quadrupole structure, which was used to cool and accumulate the incoming ions (Nieminen et al., 2001). Following this the ions were extracted as short bunches with excellent ion optical properties for the injection into the double Penning trap system JYFLTRAP (Kolhinen et al., 2004), which was used to separate ^{133m}Xe in a two-step cleaning process. The first mass-selective cleaning step was performed in the helium gas-filled purification trap. This cleaning step, with a mass resolution of about 30 ppm, could remove all other ions except ^{133}Xe , ^{133m}Xe , ^{133m}I and ^{133}I . Additionally, some traces of ^{133}Te were observed after this sideband cleaning process, described in detail in Savard et al. (1991). About 250 ms was needed to accomplish the first cleaning step.

The second cleaning cycle was performed in the gas-free precision Penning trap using ion motion excitation with a dipolar RF-electric field using Ramsey's method of time-separated oscillatory fields (Eronen et al., 2008b). The contaminating ions are excited to large radial orbits while the ion species of interest remain close to the center of the trap. The ions are then extracted back towards the gas-filled purification trap whereby the contaminants are completely removed as they hit a narrow 2 mm diaphragm located between the two Penning traps. In this work, a Ramsey ion motion excitation time pattern of 20–40–20 ms (On-Off-On) was used, providing a mass resolution of about 1 ppm, enough to separate the different nuclear states of ^{133}Xe as shown in Fig. 1. Once the bunch is re-cooled in the purification trap, the isomerically clean sample is finally released for implantation and analysis. This second purification step also needed about 250 ms to complete. Approximately 30 ions per released bunch appeared to be the maximum achievable throughput.

A quadrupole frequency scan was performed in the precision trap to diagnose the peaks of Fig. 1. This scan was made only after completion of both purification steps. When the excitation frequency matches the cyclotron frequency $v_c = 1/(2\pi) B q/m$ of the ions, the ions arrive at the detector in a shorter time. In the equation the symbol B represents the magnetic field and q and m correspond to the charge and the mass of the particle. The time-of-flight ion-cyclotron resonances (König et al., 1995; Gräff et al., 1980) for different cleaning frequencies [A–C] are shown in Fig. 2. An excitation time of 400 ms was used, resulting in a FWHM of about 2 Hz, enough to clearly identify the two states of ^{133}Xe . It is apparent from the time-of-flight resonance curves shown in Fig. 2 that the different ion species are fully separated, i.e., only a single equally deep time-of-flight minimum is observed in each case.

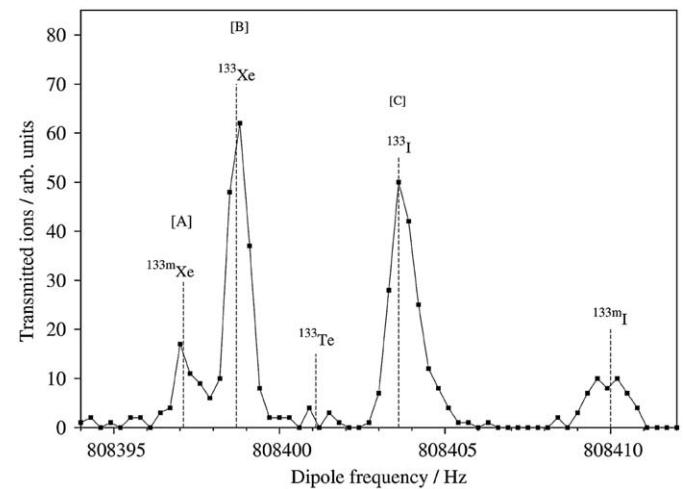


Fig. 1. Frequency scan in dipole cleaning. The number of transmitted ions is plotted as a function of the dipole frequency. The different ion species are marked. The frequencies marked with [A], [B] and [C] correspond to the transmission of ^{133m}Xe , ^{133}Xe and ^{133}I ions, respectively.

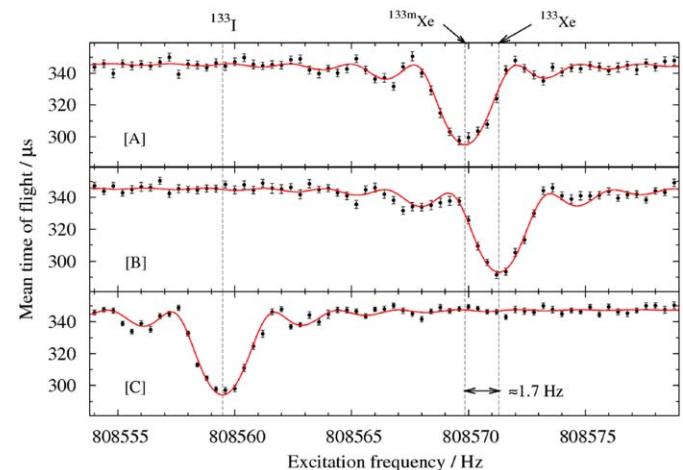


Fig. 2. Time-of-flight ion-cyclotron resonance curves for ion species that are transmitted when the dipole frequency is set to [A], [B] or [C] shown in Fig. 1. The cyclotron resonance frequencies of the three species are marked with dashed vertical lines. The relative mass difference of the states in xenon is only 2 ppm, corresponding to a cyclotron frequency difference of about 1.7 Hz.

The positions of these minima provide unambiguous identification of the two ^{133}Xe and ^{133}I peaks of Fig. 1. Based on the data presented above, the ^{133m}Xe (or ^{133}Xe) beam purity after the trap is 95%–100%.

The extracted ^{133m}Xe ions were reaccelerated to 30 keV of energy and implanted into a thin aluminum foil (thickness 3.2 mg/cm², purity 99.5%). Peräjärvi et al. (2008) demonstrates that the xenon does not rapidly escape from the aluminum foil while using such implantation settings. The typical ^{133m}Xe implantation rate was approximately 60 ions/s. With the current system performance, a sample of 10^6 atoms can be prepared in less than 5 h. A sample size of 10^5 ^{133m}Xe atoms is large enough for the calibration of the noble gas sampling instruments (Bowyer et al., 2002; Fontaine et al., 2004; Stocki et al., 2004). This corresponds to 0.37 Bq initial activity. Altogether four samples containing from 80 000 to 6.9×10^5 ^{133m}Xe atoms were successfully fabricated during two accelerator beam-time periods. The

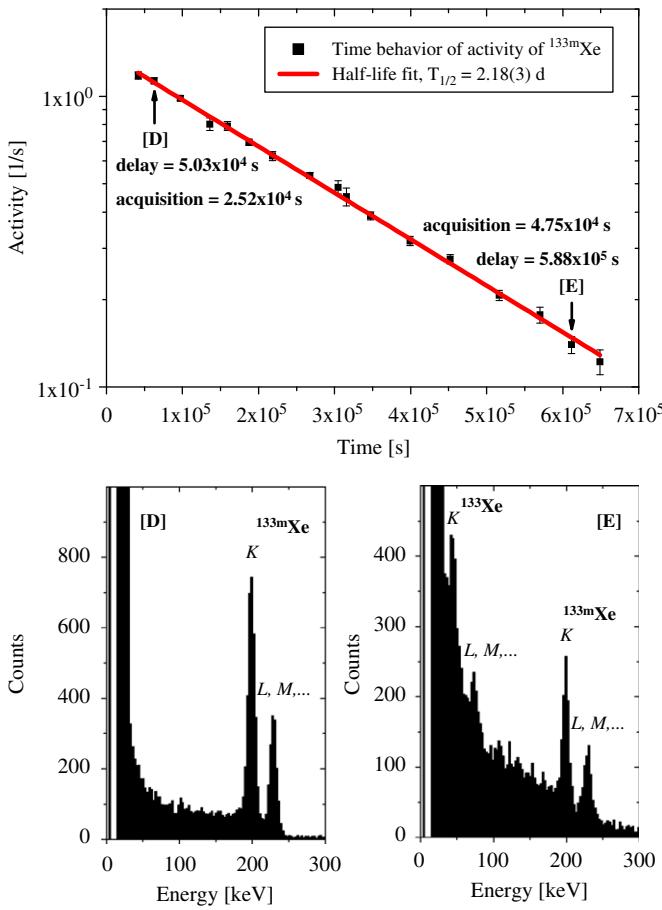


Fig. 3. Decay curve of ^{133m}Xe and conversion electron spectra. The decay curve was generated using the K shell conversion electrons. The lower part presents two electron spectra used in the generation of the upper half-life graph. The spectrum on the left belongs to the fresh sample (data point [D] in the half-life graph) and on the right for the aged sample (data point [E] in the half-life graph).

total beam-time used for the project was four days. Most of this time was reserved for the development of the above described purification process.

^{133m}Xe at an excitation energy of 233.2 keV solely decays to the more long-lived ground state of ^{133}Xe by gamma radiation or conversion electron decay (total internal conversion coefficient $\alpha=8.84(13)$) (Galán, 2009). The corresponding K- and L-shell electron energies are 198.7 and 228.0 keV, respectively. The ^{133}Xe beta-decays further to stable ^{133}Cs of which 99.9% of these decays proceed via the 81.0 keV excited state in ^{133}Cs (Galán, 2009). Conversion electrons with energies of 45.0 keV ($\alpha_K=1.429(20)$) and 75.4 keV ($\alpha_L=0.214(3)$) are often associated with the decay of this state (Kibédi et al., 2008).

Xenon remains in the aluminum matrix. This was shown by successfully determining the half-life of ^{133m}Xe using K conversion electron data from a sample initially containing 3.7×10^5 ^{133m}Xe atoms, see Fig. 3, and by baking another sample at a temperature of 610 °C in vacuum for about 1 h without losing the sample. The lower part of Fig. 3 presents two electron spectra measured with the same Si detector, one from a relatively fresh sample and the other from an aged sample. In the fresh sample there are no visible conversion electron spectrum lines below 190 keV of energy, indicating that the sample is purely ^{133m}Xe . In the aged sample however, lines with less than 100 keV of energy following the beta-decay of the ground state of ^{133}Xe are clearly seen demonstrating the growth-in of ^{133}Xe activity. By baking a further sample in air at 1000 °C for about 1.5 h, 80(20)% of xenon

was released to the gas phase. Notice that aluminum melts in air at about 660 °C. The sample also oxidized heavily during the baking. Finally, no remnants of xenon were detected after baking another sample in vacuum at 970 °C for about 1.5 h. Due to the relatively high temperatures involved performance of other catcher materials, such as graphite, will be investigated in the future. The search of a catcher material with more favorable xenon release behavior is important for the production of gaseous xenon samples. If desired, pure ^{131m}Xe and ^{135}Xe sources can also be fabricated using the developed method.

The introduced technique can be expanded in such a way that the implantation time of every single ion is recorded. This means that the implantation time profiles can be constructed and the exact number of implanted ions can be calculated. Such on-line monitoring would become possible by designing a setup where the implantation foil is tilted and viewed with a channeltron or a MCP detector which can detect the electrons that are released from the foil while ions are being implanted. Since the time resolution of a channeltron or a MCP detector is around 20 ns and the length of the ion pulse about 10 μs , all the ions can be effectively counted and thus an accurate number of implanted ions can be deduced.

3. Discussion

Noble gas samplers and spectrometers deployed around the globe are continuously operated by the CTBTO. The demonstrated ability to produce ultra-pure xenon samples potentially is of immense value to the work of this organization. Noble gas measuring systems need to be calibrated using single xenon isotopes and isomers; quality control and intercomparison exercises between CTBT laboratories are possible applications. Standardization of the detection capability of xenon detectors is crucial for producing reliable and accurate results for verification of the CTBT.

Additionally, since the developed purification process is fast (typically 500 ms), the present work opens up other interesting prospects both to basic and applied fields either in the form of ultra-pure samples of unstable nuclei or possibly as an improved analytical tool to accurately determine the yields of various nuclear reaction products. Accurate knowledge of neutron-induced independent fission yields would, for example, be very valuable for the development of Generation IV gas-cooled fast reactors for sustainable energy production (Bosq et al., 2006) and for the development and testing of nuclear fission theory.

Acknowledgements

This work was supported in part by the Academy of Finland under the Finnish Centre of Excellence Program 2006–2011 (Nuclear and Accelerator Based Physics Program at JYFL).

References

- Äystö, J., 2001. Development and applications of the IGISOL technique. *Nucl. Phys. A* 693, 477–494.
- Bosq, J.C., Brun-Magaud, V., Rimpault, G., Tommasi, J., Conti, A., Garnier, J.C., 2006. GFR core neutronics studies at CEA. In: Rullhusen, P. (Ed.), *Proceedings of the International Workshop on Nuclear Data Needs For Generation IV Nuclear Energy Systems*, Antwerpen, Belgium, 5–7 April, 2005. World Scientific Publishing, Singapore, pp. 46–57.
- Bowyer, T.W., Schlosser, C., Abel, K.H., Auer, M., Hayes, J.C., Heimbigner, T.R., McIntyre, J.I., Panisko, M.E., Reeder, P.L., Satorius, H., Schulze, J., Weiss, W., 2002. Detection and analysis of xenon isotopes for the comprehensive nuclear-test-ban treaty international monitoring system. *J. Environ. Radioact.* 59, 139–151.

Brown, L.S., Gabrielse, G., 1986. Geonium theory: physics of a single electron or ion in a penning trap. *Rev. Mod. Phys.* 58, 233–311.

Eronen, T., Elomaa, V.-V., Hager, U., Hakala, J., Hardy, J.C., Jokinen, A., Kankainen, A., Moore, I.D., Penttilä, H., Rahaman, S., Rinta-Antila, S., Rissanen, J., Saastamoinen, A., Sonoda, T., Weber, C., Äystö, J., 2008a. Q_{EC} values of the superallowed β emitters ^{50}Mn and ^{54}Co . *Phys. Rev. Lett.* 100, 132502.

Eronen, T., Elomaa, V.-V., Hager, U., Hakala, J., Jokinen, A., Kankainen, A., Rahaman, S., Rissanen, J., Weber, C., Äystö, J., 2008b. Preparing isomerically pure beams of short-lived nuclei at JYFLTRAP. *Nucl. Instrum. Methods Phys. Res. B* 266, 4527–4531.

Fontaine, J.-P., Pointurier, F., Blanchard, X., Taffary, T., 2004. Atmospheric xenon radioactive isotope monitoring. *J. Environ. Radioact.* 72, 129–135.

Galán, M., 2009. Evaluation of ^{133}Xe and $^{133}\text{Xe}^m$ decay data. *Appl. Radiat. Isot.* 67, 170–173.

Gräff, G., Kalinowsky, H., Traut, J., 1980. A direct determination of the proton-electron mass ratio. *Z. Phys. A* 297, 35–39.

Kibédi, T., Burrows, T.W., Trzhaskovskaya, M.B., Davidson, P.M., Nestor Jr., C.W., 2008. Evaluation of theoretical conversion coefficients using BrIcc. *Nucl. Instrum. Methods Phys. Res. A* 589, 202–229.

Kolhinen, V.S., Kopecky, S., Eronen, T., Hager, U., Hakala, J., Huikari, J., Jokinen, A., Nieminen, A., Rinta-Antila, S., Szerypo, J., Äystö, J., 2004. JYFLTRAP: a cylindrical Penning trap for isobaric beam purification at IGISOL. *Nucl. Instrum. Methods Phys. Res. A* 528, 776–787.

König, M., Bollen, G., Kluge, H.-J., Otto, T., Szerypo, J., 1995. Quadrupole excitation of stored ion motion at the true cyclotron frequency. *Int. J. Mass Spectrom. Ion Processes* 142, 95–116.

New Scientist, 2007. Nuke sniffer, vol. 196, No. 2627, 4.

Nieminen, A., Huikari, J., Jokinen, A., Äystö, J., Campbell, P., Cochrane, E.C.A., 2001. Beam cooler for low-energy radioactive ions. *Nucl. Instrum. Methods Phys. Res. A* 469, 244–253.

Peräjärvi, K., Turunen, J., Hakala, J., Jokinen, A., Moore, I.D., Penttilä, H., Saastamoinen, A., Siiskonen, T., Toivonen, H., Äystö, J., 2008. The decay of ^{133m}Xe . *Appl. Radiat. Isot.* 66, 530–534.

Reeder, P.L., Bowyer, T.W., 1998. Delayed coincidence technique for ^{133g}Xe detection. *Nucl. Instrum. Methods Phys. Res. A* 408, 573–581.

Ringbom, A., Elmgren, K., Lindh, K., Peterson, J., Bowyer, T., Hayes, J.C., McIntyre, J.I., Panisko, M., Williams, R., 2009. Measurements of radioxenon in ground level air in South Korea following the claimed nuclear test in North Korea on October 9, 2006. In: International Scientific Studies Conference, Hofburg Imperial Palace, Vienna, Austria, 10–12 June, 2009. Abstract book, pp. 93.

Savard, G., Becker, St., Bollen, G., Kluge, H.-J., Moore, R.B., Otto, Th., Schweikhard, L., Stolzenberg, H., Wiess, U., 1991. A new cooling technique for heavy ions in a penning trap. *Phys. Lett. A* 158, 247–252.

Stocki, T.J., Bean, M., Ungar, R.K., Toivonen, H., Zhang, W., Whyte, J., Meyerhof, D., 2004. Low level noble gas measurements in the field and laboratory in support of the Comprehensive Nuclear-Test-Ban Treaty. *Appl. Radiat. Isot.* 61, 231–235.

Wernsperger, B., Schlosser, C., 2004. Noble gas monitoring within the international monitoring system of the comprehensive nuclear test-ban treaty. *Radiat. Phys. Chem.* 71, 775–779.